

Giant Anisotropy of Magnetoresistance and "Spin Valve" effect in Antiferromagnetic $Nd_{2-x}Ce_xCuO_4$

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We have studied anisotropic magnetoresistance (MR) and magnetization with rotating magnetic field (B) within CuO_2 plane in lightly doped AF $Nd_{2-x}Ce_xCuO_4$. A giant anisotropy in MR is observed at low temperature below 5 K. The c-axis resistivity can be tuned about one order of magnitude just by changing B direction within CuO_2 plane and a scaling behavior between out-of-plane and in-plane MR is found. A "Spin valve" effect is proposed to understand the giant anisotropy of out-of-plane MR and the evolution of scaling parameters with the external field. It is found that the field-induced spin-flop transition of Nd^{3+} layer under high magnetic field is the key to understand the giant anisotropy. These results suggest that a novel entanglement between charge and spin dominates the underlying physics.

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I. INTRODUCTION

It is generally believed that the pairing necessary for high- T_c superconductivity in cuprates involves the interplay between doped charges and antiferromagnetic (AF) spin correlation. In this sense, the study of lightly doped, insulating AF state is important to understand the pairing mechanism because density of the carriers can be sufficiently low so that the interaction between them is small relative to their interaction with Cu^{2+} spins. Many intriguing and anomalous phenomena were observed in lightly doped AF cuprates due to strong coupling between charges and Cu^{2+} spins.^{1,2,3,4,5} Cu^{2+} spins order in an AF collinear structure for the parent compounds of hole-doped cuprates,^{6,7} while in AF noncollinear structure for that of electron-doped cuprates.^{8,9} All spins point either parallel or antiparallel to a single direction in AF collinear structure, while the spins in adjacent layers are orthogonal in AF noncollinear structure. A transition from noncollinear to a collinear spin arrangement with a spin-flop can be induced by certain magnetic field (B_c)¹⁰, which is confirmed in lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ ⁴ and $Nd_{2-x}Ce_xCuO_4$ ⁵ crystals, and such transition affects significantly both the in-plane and out-of-plane resistivity.

In Nd_2CuO_4 , the Cu^{2+} spins order in three phases with two different AF noncollinear spin structures and experience two reorientation phase transitions^{8,11,12,13}. It has been reported by us that MR anisotropy with a fourfold symmetry in different AF spin structures upon rotating magnetic field (B) within ab-plane, while with a twofold symmetry at the spin reorientation temperatures, is observed in lightly doped $Nd_{2-x}Ce_xCuO_4$ above 10 K.⁵ A large anisotropic MR was observed in lightly electron-doped $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ ⁴ and $Nd_{2-x}Ce_xCuO_4$ ⁵. These results indicate strong spin-charge coupling in electron-doped cuprates. In

Nd_2CuO_4 , the magnetic coupling between Nd^{3+} and Cu^{2+} is very important at low temperature since the magnetic moment of Nd^{3+} becomes large with decreasing temperature ($\sim 1.3\mu_B$ at 0.4 K).¹² Magnetic structure of Nd^{3+} is very abundant at low temperature.^{14,15,16} In this sense, electronic transport at low temperature is expected to be sensitive for the change of magnetic structure of Nd^{3+} due to strong spin-charge coupling. It will provide us a chance to understand the spin-charge coupling in electron-doped cuprates. The lightly electron-doped cuprates are good system to study the coupling between charge and Cu^{2+} spin because: (1) the spin structure can be tuned by external magnetic field;¹⁰ (2) in contrast to the buckling of CuO_2 in hole doped cuprates, the CuO_2 plane in electron-doped cuprates is flat, so that the spin ordering is pure antiferromagnetic without ferromagnetic component along c-axis occurred in hole doped cuprates, such ferromagnetic component along c-axis makes the study of the coupling between charge and Cu^{2+} spin complicated. In this work, we study angular dependent magnetoresistance and magnetization below 10 K in lightly electron-doped $Nd_{2-x}Ce_xCuO_4$. A giant anisotropy in MR is observed, and the c-axis resistivity can be tuned about one order of magnitude just by changing B direction. Scaling behavior between in-plane and out-of-plane MR is systematically changed with increasing magnetic field. The jump in MR with B around Cu-O-Cu direction coincides with the sudden change in magnetization at low temperature below 5 K. The underlying physics will be discussed below.

II. EXPERIMENT

Growth of single crystals and their resistivity have been reported in previous work.⁵ Susceptibility and magnetoresistance were measured with the superconducting

quantum interference device (SQUID) with 7 Tesla maximal magnetic field and quantum design PPMS system with 12 Tesla maximal magnetic field, respectively. In our measurements, the maximal magnetic field is 12 Tesla for MR and 7 Tesla for magnetization. The ρ_{ab} and ρ_c stand for in-plane resistivity and out-of-plane resistivity, respectively. The magnetoresistance is defined as $MR = \frac{\Delta\rho(B)}{\rho(0)} = \frac{\rho(B) - \rho(0)}{\rho(0)}$. It should be addressed that all results discussed as follow are well reproducible.

III. RESULT AND DISCUSSION

Fig.1 shows the isothermal out-of-plane MR at 5 K for the single crystals with $x=0.025$ and 0.033 with B along Cu-Cu and Cu-O-Cu direction, respectively. The MR behavior is similar to that observed in antiferromagnetic $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with $x=0.01$ crystal.⁴ But the magnitude of MR and the MR anisotropy are much larger than the case of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$. The step-like increase of MR corresponds to the noncollinear-collinear transition occurring at the critical field B_c . As shown in Fig.1, the critical field B_c along Cu-O-Cu direction is larger than that along Cu-Cu direction. Above B_c , the behavior of MR for B along Cu-Cu direction is quite different from that for B along Cu-O-Cu direction in the collinear structure. The MR with B along Cu-Cu direction slightly changes above B_c , while the MR monotonically increases with increasing B for B along Cu-O-Cu direction. A giant anisotropic MR between B along Cu-Cu and Cu-O-Cu direction is observed. For $x=0.025$ crystal, the MR at 12 T is as high as $\sim 235\%$ with B along Cu-O-Cu direction, while only $\sim 17\%$ with B along Cu-Cu direction.

Upon rotating a magnetic field larger than B_c within the CuO_2 plane, the spins always keep the collinear arrangement and the spin structure rotates as a whole, all spins are perpendicular to the magnetic field as shown in Fig.1.¹⁰ In order to study the anomalous and giant anisotropic MR, we carefully investigated the evolution of in-plane and out-of-plane MR with rotating B within CuO_2 plane. Fig. 2a and 2b show the evolution of in-plane and out-of-plane MR with the angle between B and Cu-O-Cu ([100]) direction at 5 K for the single crystal with $x=0.025$. Both of in-plane and out-of-plane MR increase with increasing B , and show a giant anisotropy with fourfold-symmetry, such fourfold symmetry arises from the symmetry of magnetic structure because there exist two equivalent spin easy axes (Cu-Cu direction) and two equivalent spin hard axes (Cu-O-Cu direction) in the collinear spin structure, which has been confirmed by the different critical fields B_c for B along Cu-O-Cu and Cu-Cu directions as shown in Fig.1. A striking feature is observed that the out-of-plane MR at 12 T sharply increases from $\sim 200\%$ to $\sim 300\%$ at the angle close to B along Cu-O-Cu. Such behavior originates from the spin-flop induced by magnetic field with B close to the Cu-O-Cu direction as discussed below. A similar jump

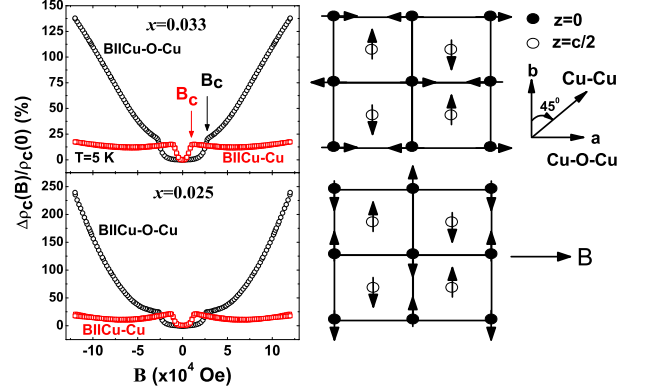


FIG. 1: Isothermal MR at 5 K with the B along Cu-O-Cu and Cu-Cu direction for the samples $Nd_{2-x}Ce_xCuO_4$ with $x=0.025$ and 0.033 , respectively. Zero-field noncollinear spin structure, only Cu spins are shown; Field-induced transition from noncollinear to collinear spin ordering with B along Cu-O-Cu direction.

can be also observed at the angle close to B along Cu-Cu, but the jump is very small compared to the case of B close to Cu-O-Cu direction.

In order to study the effect of temperature on the anisotropy of MR, we systematically investigated the MR behavior of the $x=0.033$ crystal because resistivity of the $x=0.025$ crystal is too large to be measured due to the resistivity divergence at low temperature. Fig.3 shows evolution of the out-of-plane MR upon rotating B within CuO_2 plane at 2, 4 and 5 K under 12 T for the $x=0.033$ crystal. The results are similar to that observed in the crystal with $x=0.025$. The MR increases monotonically and the anisotropy of MR induced by rotating B within Cu-O plane apparently increases with decreasing temperature. The MR under 12 T with B along Cu-Cu direction is about 11.2% at 5 K, 17.1% at 4 K and 27.7% at 2 K; while the MR with B along Cu-O-Cu direction is about 133% at 5 K, 203% at 4 K and 656% at 2 K, respectively. It indicates that a giant anisotropy of resistivity is induced by magnetic field with B along Cu-O-Cu and Cu-Cu at low temperature. At 2 K, the resistivity under 12 T with B along Cu-O-Cu direction is about one order of magnitude larger than that with B along Cu-Cu direction. Such giant anisotropy in resistivity induced just by changing B direction within CuO_2 plane should be related to the magnetic structure and magnetic moment induced by B because the magnetic field along Cu-O-Cu or Cu-Cu just changes the spin structure and induces the different magnitude of the magnetic moment. To understand the jump in MR with B close to the Cu-O-Cu direction, the MR at 5 K is measured with rotating B in clockwise direction and in anti-clockwise direction, respectively. It is found that the MR jumps observed with rotating B in clockwise direction and in anti-clockwise

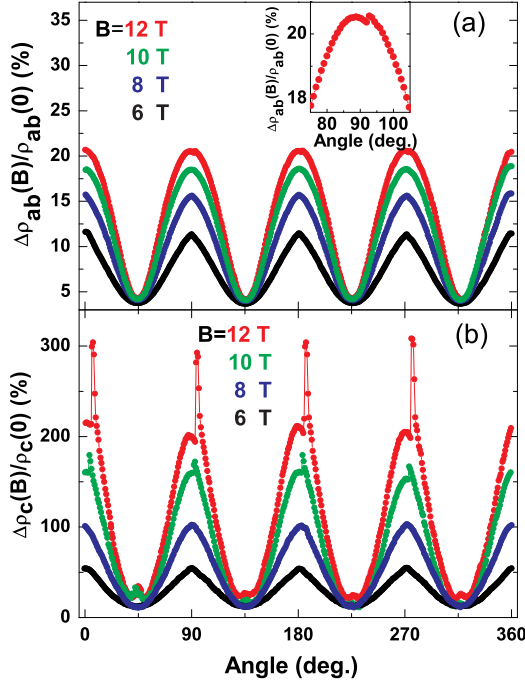


FIG. 2: (a): Isothermal in-plane and (b): Out-of-plane MR at 5 K under different B as a function of angle between B and Cu-O-Cu direction upon rotating B within CuO_2 plane for the single crystal with $x=0.025$. The inset in (a): magnified in-plane MR with $B = 12$ T.

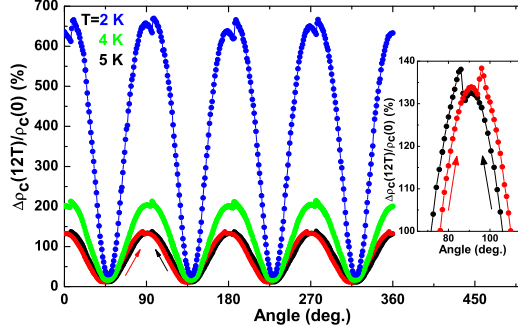


FIG. 3: Out-of-plane MR as a function of angle between B and Cu-O-Cu direction upon rotating B within CuO_2 plane at 2K, 4K and 5 K, respectively, for the single crystal with $x=0.033$ ($B=12$ T).

direction are symmetric relative to the B along Cu-O-Cu direction as shown in the inset of Fig.3. It indicates that the spin does not prefer to the Cu-O-Cu direction, and the spin jump always occurs around Cu-O-Cu direction when B is rotated within CuO_2 plane. Therefore, the jump arises from the spin-flop induced by B.

As shown in Fig.4, the same data of in-plane and out-of-plane MR shown in Fig.2a and 2b are plotted in $\Delta\rho_c(B)/\rho_c(0)$ as a function of $\Delta\rho_{ab}(B)/\rho_{ab}(0)$. Only the

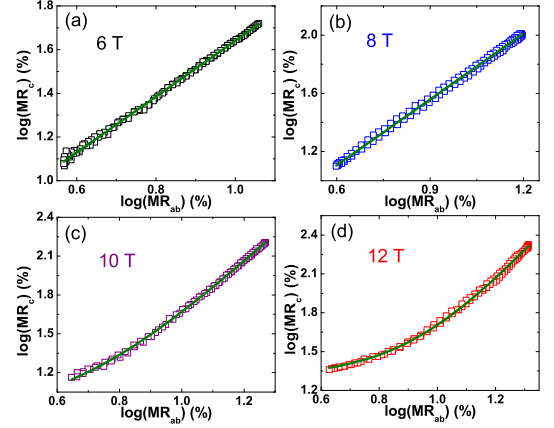


FIG. 4: The same data shown in Fig.2 are plotted in $\Delta\rho_c(B)/\rho_c(0)$ as a function of $\Delta\rho_{ab}(B)/\rho_{ab}(0)$. The line is the fitting results with the formula $\Delta\rho_c(B)/\rho_c(0) = \beta + \alpha(\Delta\rho_{ab}(B)/\rho_{ab}(0))^v$. At 6 T and 8 T, the parameter $\beta = 0$.

TABLE I: Fitting parameters α , β and v with the formula $\Delta\rho_c(B)/\rho_c(0) = \beta + \alpha(\Delta\rho_{ab}(B)/\rho_{ab}(0))^v$ under different field.

Field	β	α	v
6 T	0	2.30	1.29
8 T	0	1.59	1.51
10 T	7.47	0.25	2.20
12 T	20.13	0.10	2.50

data of in-plane and out-of-plane MR between 45 and 90 degree is plotted in Fig.4 because in-plane and out-of-plane MR exhibit the exactly same oscillation. All data above can be fitted by $MR_c = \beta + \alpha \cdot MR_{ab}^v$ very well. The fitting parameters are list in Table I. It is found that the fitting parameter β is zero below 10 T. The fitting parameter v increases from ~ 1 to ~ 3 with increasing magnetic field. These results indicate that the relation between out-of-plane MR and in-plane MR is strongly dependent on the external magnetic field. It is suggested that the out-of-plane and in-plane transport is closely related to magnetic structure since the external magnetic field can modify the spin structure. The giant anisotropy could arise from the change of spin structure induced by external magnetic field.

In order to further understand how magnetic field influences the transport, understanding on the evolution of magnetic structure under magnetic field is very necessary. Fig.5 shows the magnetization under 7 Tesla at 2 K with rotating B within CuO_2 plane for the AF $Nd_{2-x}Ce_xCuO_2$ with $x=0, 0.025, 0.06$ and 0.13 . It is found that magnetization shows the same fourfold symmetry with rotating B within CuO_2 plane as that observed in MR shown in Fig.2. The amplitude of the oscillation and the magnetization decrease with increasing x . A striking feature is observed that the magnetization

shows a jump with B around Cu-O-Cu direction at which a corresponding jump is observed in MR as shown in the inset of Fig.5. However, this fourfold symmetry gradually disappears with increasing temperature as shown in Fig.6. As we know, the magnetic moment of Nd^{3+} increases prominently at low temperature, and the magnetization is very sensitive to magnetic moment of Nd^{3+} below 5 K^{14,17}. Therefore, the fourfold symmetry in magnetization and the jump in magnetization are related to the magnetic structure of Nd^{3+} . In the other hand, the fourfold symmetry shown in Fig.5 indicates a magnetic ordering of Nd^{3+} . The spontaneous ordering of the Nd^{3+} subsystem at low temperature due to Nd^{3+} - Nd^{3+} interaction remains controversial. X-ray magnetic scattering data indicate that Nd^{3+} ions are polarized at 37 K¹⁸. The removal of the Kramers doublet degeneracies observed by crystal field infrared transmission indicates that these ions are already polarized by the Cu^{2+} subsystem at a temperature as high as 140 K¹⁹. An enhancement of neutron scattering magnetic peak intensities around 3 K has been interpreted as Nd^{3+} ordering due to Nd^{3+} - Nd^{3+} interaction¹², while Lynn et al. have estimated the Nd^{3+} ordering temperature around 1.5 K¹⁷. Recently, an abnormal peak around 5 K observed in ultrasonic measurement is explained to be somehow related to local magnetic domains¹⁵. Since the Nd^{3+} - Cu^{2+} and Nd^{3+} - Nd^{3+} interactions are opposite, the former is dominated above 5 K and makes Nd^{3+} parallel to Cu^{2+} as shown in Fig.8(a), while the later is dominated below 5 K and makes Nd^{3+} prefer to be perpendicular to Cu^{2+} as shown in Fig.8(b). Due to the frustration of the Nd^{3+} magnetic subsystem arising from the competition between Nd^{3+} - Cu^{2+} and Nd^{3+} - Nd^{3+} interaction, the local magnetic domain is formed with Nd^{3+} not parallel to Cu^{2+}

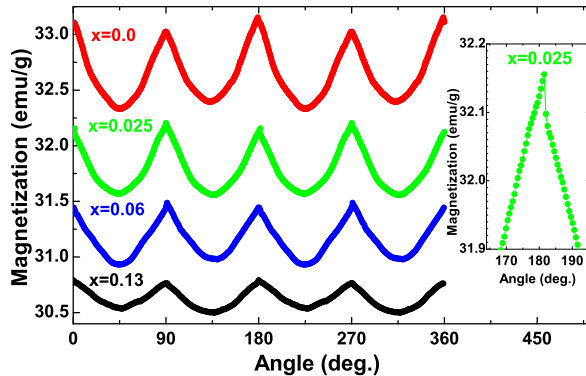


FIG. 5: The magnetization measured under magnetic field of 7 Tesla at 2 K as a function of angle between B and Cu-O-Cu direction upon rotating B within CuO_2 plane for the samples with $x=0, 0.025, 0.06, 0.13$. Inset shows a jump in magnetization at certain angle corresponding to the jump observed in MR.

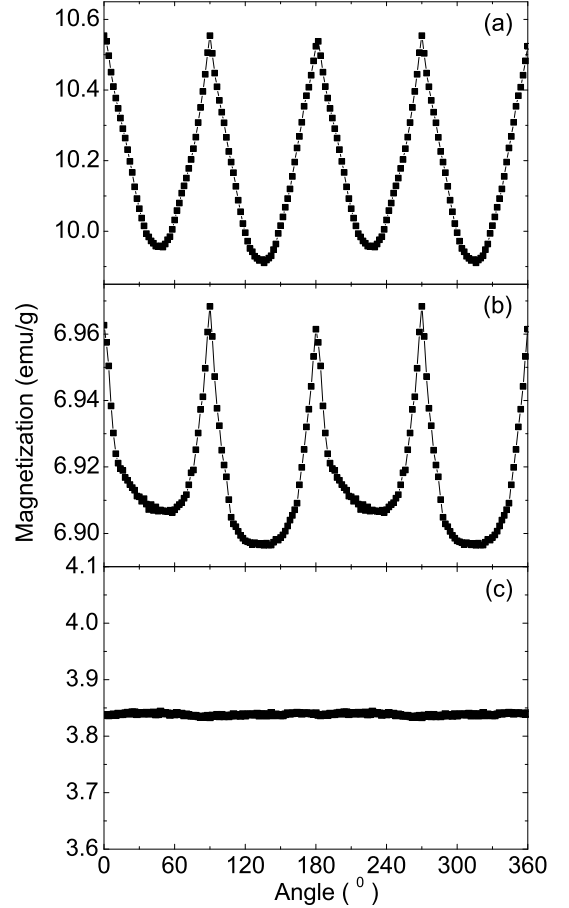


FIG. 6: The increment of magnetization measured under magnetic field of 6.5 Tesla at (a): 2 K, (b): 3.5 K and (c): 5 K relative to the magnetization at 7.5 K as a function of angle between B and Cu-O-Cu direction upon rotating B within CuO_2 plane for the samples with $x=0.025$.

magnetic moment below 5 K. The magnetic structure of Nd^{3+} subsystem with magnetic moment of Nd^{3+} perpendicular to Cu^{2+} can be stabilized by the external field. The observed fourfold symmetry below 5K in magnetization could arise from the reorientation of Nd^{3+} spin. Richard et al. have given evidence that the magnetic structure below 5 K has anisotropic field-dependence¹⁵. As shown in Fig.7, the field dependent magnetization of $x=0.025$ sample at 2 K shows an anomaly at 0.6 T and 3.6 T for both Cu-Cu and Cu-O-Cu directions, respectively. However, no such anisotropy is observed above 5 K. To make the anisotropy clear, the magnetization at 2 K subtracted the 5 K magnetization is shown in Fig.7(b). Such anomaly has been attributed to spin-reorientation of Nd^{3+} in Nd_2CuO_4 ¹⁴. The spin reorientation of Nd^{3+} occurs due to a transition from the magnetic structure shown in Fig.8(a) to that shown in Fig.8(b). It is surprising that the magnetization for two directions has a cross around 6 T and the magnetization shows somehow saturation as shown in Fig. 7. It is suggested that the

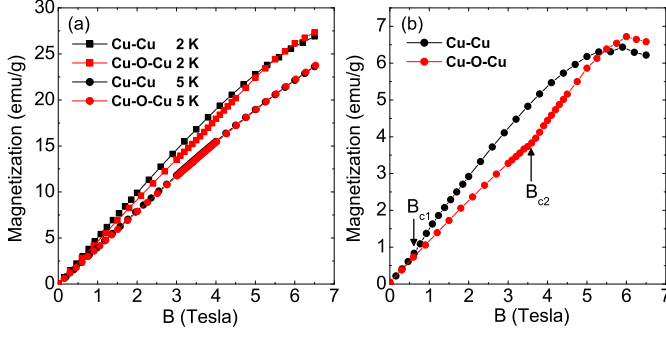


FIG. 7: (Color online) (a): Magnetization as a function of magnetic field along Cu-Cu and Cu-O-Cu direction at 2 K and 5 K; (b): $M(2K)-M(5K)$.

magnetic structure under high magnetic field is different from that under low magnetic field. Similar result has been reported in Nd_2CuO_4 ¹⁴. Recently, a crossover from antiferromagnetic to paramagnetic configuration induced by high magnetic field is proposed by Richard et al.¹⁶. The corresponding magnetic structures given by Richard et al. are shown in Fig.8(c)-(f). When in-plane magnetic field $B < 4$ T along Cu-O-Cu and Cu-Cu directions, the collinear magnetic structures are shown in Fig.8(c) and (e), respectively. In the configuration, the Cu^{2+} - Nd^{3+} interaction is larger than Nd^{3+} - Nd^{3+} interaction. When in-plane magnetic field $B > 4$ T along Cu-O-Cu and Cu-Cu directions, the Nd^{3+} - Nd^{3+} interaction is dominated and the magnetic structure changes from antiferromagnetic to paramagnetic configuration as shown in Fig.8 (d) and (f), in which the Nd^{3+} spins are aligned in applied magnetic field and thus behave as ferromagnetic-like. The cross at about 6 T in magnetization could be related to the change of magnetic structure shown in Fig.8.

The fourfold symmetry in magnetoresistance has been observed above 5 K in the same sample in previous result⁵, while similar symmetry in magnetization arose from Nd^{3+} spin is observed only below 5 K. Therefore, the fourfold symmetry in MR should arise from anisotropic magnetic structure of Cu^{2+} as discussed in our previous work⁵. As shown in Fig.2 and Fig.6, the giant anisotropy in MR below 5 K coincides with the magnetic ordering of Nd^{3+} with the same fourfold symmetry. These results indicate that the fourfold symmetry of MR results from spin ordering of Cu^{2+} , and the spin ordering of Nd^{3+} enhances the fourfold symmetry in MR below 5 K and leads to a giant anisotropy in MR. It is evident that the jump of out-of-plane MR with B along Cu-O-Cu direction shown in Fig.2 is related to the sudden change in magnetization shown in the inset of Fig.5. Therefore, the change of MR below 5 K relative to high temperature MR can be mainly ascribed to the ordering of Nd^{3+} spin. As shown in Fig.4, there exists a scaling behavior between the out-of-plane MR and in-plane MR with $\text{MR}_c = \beta + \alpha \cdot \text{MR}_{ab}^v$. But the fitting parameters

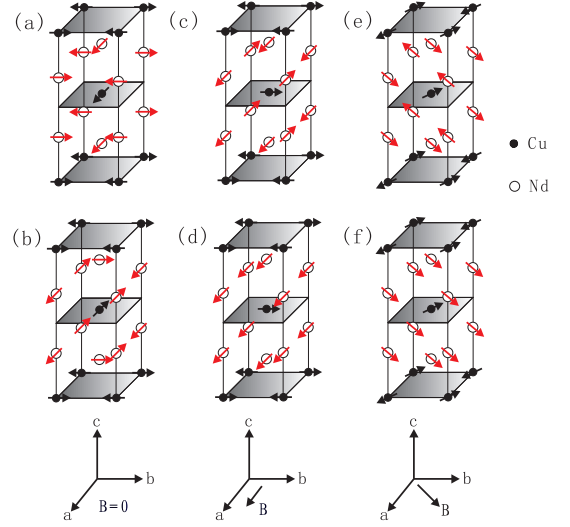


FIG. 8: (Color online) Magnetic configuration of Nd_2CuO_4 in the noncollinear antiferromagnetic phase (a): above 5 K; (b): below 5 K; Magnetic configuration of Nd_2CuO_4 in the collinear phase assuming an antiferromagnetic alignment of Nd^{3+} spins (c): $B \parallel [100]$; (e): $B \parallel [110]$, and a ferromagnetic-like alignment of the Nd^{3+} spins (d): $B \parallel [100]$; (f): $B \parallel [110]$.

β , α and v show a systematic change with increasing external field as listed in Table 1. Change of the scaling behavior is closely related to the change of magnetic structure of Nd^{3+} induced by external field since the increase of external field from 6 T to 12 T cannot lead to change of magnetic structure of Cu^{2+} subsystem. It is supported by the fact that the change of in-plane MR with increasing magnetic field is much smaller than that of out-of-plane MR, and the anisotropy of out-of-plane MR is much larger than that of in-plane MR as shown in Fig.2. Richard et al.¹⁶ pointed out that the magnetic structure of Nd^{3+} subsystem can change from antiferromagnetic to paramagnetic configuration at certain critical magnetic field. It is possible that the evolution of scaling parameters listed in Table 1 is related to the change of magnetic structure of Nd^{3+} . It is well known that the neighboring CuO_2 plane with antiferromagnetic configuration is separated by Nd-O layer.¹⁵ As discussed above, the magnetic structure of Nd^{3+} subsystem can be turned by external field. Therefore, the out-of-plane transport can be switched by magnetic field assuming the Nd-O layer as a barrier. In this sense, this phenomenon can be well understood with "Spin Valve" effect. The magnetic excitations are different with B along Cu-Cu and Cu-O-Cu directions because the magnetic structure is more frustrated around Cu-O-Cu¹⁵. In "Spin Valve" picture, the different magnetic excitations in Nd^{3+} layer lead to different transport along c-axis. Therefore, the spin-flop transition is the key to understand the giant anisotropy of out-of-plane MR. Thermal conductivity results indicate that in-plane magnetic field can result in a close of anisotropic gap ($\sim 0.3\text{meV}$) which leads to ad-

ditional heat conduction²⁰. The critical magnetic field is about 4.5 T and 2.5 T in Cu-O-Cu and Cu-Cu direction, respectively. The closure of anisotropic gap corresponds to spin-flop transition for Cu^{2+} spin. This result indicates that the spin-flop transition can lead to a closure of anisotropic gap. At low temperature below 5 K, another gap related to spin-flop transition of Nd^{3+} is closed under high magnetic field and this gap is anisotropic. The gap can be estimated with $B=\Delta/g\mu_B$ ($B \sim 8T$), the gap is about 0.5 meV with magnetic field along Cu-O-Cu direction. This spin-flop should originate from Nd^{3+} subsystem because magnons from Cu^{2+} have energy above 5 meV^{21,22,23}, and four optical Nd magnon branches lie in the range 0.2 to 0.8 meV^{24,25}. The study on magnetic structure under high magnetic field at low temperature is lacking. This picture needs further experimental investigation to confirm. These results give a strong evidence that a nontrivial correlation between charge and AF ordering background exists, the charge transport can be affected not only by Cu^{2+} spins but also Nd^{3+} spins, especially below 5 K.

The huge changes in resistivities as induced by the in-plane magnetic field seem to be a highly nontrivial phenomenon. Note that the applied in-plane magnetic field should only affect the spins of the system via the Zeeman effect without directly influencing the orbital motion of charge carriers in the in-plane case, and presumably with only a weak orbital effect for the out-of-plane case as the resistivity itself is divergent at low temperature. It thus implies the existence of some kind of strong “entanglement” between the spin and charge degrees of freedom such that by tuning the magnetic ordering with an in-plane magnetic field can result in a big enhancement of resistivities seen in the measurements. Furthermore, the large MR behavior in this insulating regime also strongly suggests that the divergence of resistivities at low-temperature may not be simply a conventional localization effect due to disorders since spin structures can affect resistivities so much. Although the microscopic mechanism remains unclear, the novel spin-charge entanglement does exist in strongly correlated models. For example, in the t-J model, a so-called phase string effect has

been shown²⁶ to be present as a non-local mutual frustration between the charge and spin degrees of freedom induced by doped charge carriers moving in an antiferromagnet. In fact, the localization of the charge carriers in the magnetic ordered phase has been interpreted²⁷ based on such a phase string effect and it is thus conceivable that the change of the spin structure may strongly affect the resistivities via the phase string effect. The scaling between in-plane and out-of-plane MR is strongly dependent on spin structure of Nd^{3+} which emerges at low temperature since strong Cu^{2+} - Nd^{3+} interaction and can be easily tuned by external magnetic field. It provides a good chance to show a evidence for the spin-charge entanglement.

IV. CONCLUSION

In this paper, we study anisotropic magnetoresistance (MR) and magnetization with rotating magnetic field (B) within CuO_2 plane in lightly doped AF $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$. A giant anisotropy in MR is observed, and the c-axis resistivity can be tuned by about one order of magnitude just with changing B direction. These results provide evidence to support the spin-flop transition of Nd^{3+} ions induced by high magnetic field. The change of magnetic structure induced by different external field leads to a systematic evolution of the scaling behavior between in-plane and out-of-plane MR. “Spin Valve” effect is used to well understand the out-of-plane MR behavior. Such novel entanglement of charge and spin dominates the underlying physics.

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